Fully-tunable femtosecond laser source in the ultraviolet spectral range

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Abstract

We demonstrate experimentally the full tunability of a coherent femtosecond source in the whole ultraviolet spectral region. The experiment relies on the technique of high-order harmonic generation driven by a near-infrared parametric laser source in krypton gas. By tuning the drive wavelength in the range between 1100 to 1900 nm, we generated intense harmonics from near to extreme ultraviolet. A number of photons per shot of the order of 10^7 has been measured for the first harmonic orders. Many novel scientific prospects are expected to benefit from the use of such a table-top tunable source.

1. Introduction

Generating tunable sub-picosecond radiation at wavelengths shorter than 250 nm is of great interest to many applications in physics, chemistry and biology [1, 2, 3], both in gas-phase and condensed matter. In fact, below such a wavelength one overcomes the work function of most solids and clusters of metals and metalloids, making it possible to eject electrons from the target sample via optical excitation. The use of short pulses allows one to obtain important information on the dynamics of fast processes occurring in systems such as proteins, enzymes and nucleic acids [4]. Such a dynamics is often excited in a given small range of wavelengths (typically few nanometers). Hence, "fine" tunability is an important asset of the employed radiation source. A more extended tunability (tens of nanometers) is of course also highly desirable, since it allows the study of a large variety of samples, with the same radiation source.

Wavelengths below 150 nm are not accessible to conventional lasers, due to the lack of high-reflectivity broadband mirrors and to the decreasing efficiency of harmonic conversion in nonlinear crystals within this spectral range. Synchrotron radiation is not a viable option either, mainly because pulse duration is generally limited to several picoseconds. It should be noted that slicing schemes allow the production of sub-picosecond pulses [5], at the cost of a greatly reduced number of photons. Apart from free-electron lasers [6, 7], the most effective way to obtain femtosecond coherent pulses in the ultraviolet and soft X-ray spectral region relies on the technique of high-order harmonic generation (HHG) in rare gases [8, 9, 10]. The latter is a highly nonlinear process based on the interaction of a noble gas with a visible or infrared (IR) drive laser beam spatially focused so as to reach an intensity of the order of 10^{14} W/cm^2. The outcome of such an interaction is the generation of a comb of odd harmonics of the laser wavelength. For the first few harmonic orders (third, fifth, seventh), the signal intensity falls with the harmonic order and the process is well explained in the frame of classical nonlinear optics in centro-symmetric media [11, 12]. Higher-order harmonics instead have almost the same intensity, thus forming a characteristic plateau. The photon energy at which the plateau's cutoff is located is proportional to the square of the fundamental wavelength. The HHG process can be explained by means of the quantum model reported in [13]. However, the main features of the harmonic emission can be accounted for using a semiclassical model [14, 15].

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Generally, the drive beam for HHG has a fixed fundamental wavelength, most often around 800 nm for a standard Ti:Sapphire (Ti:Sa) source. Thus, harmonic wavelengths are also fixed. Because harmonic peaks are far away from one another, especially for low harmonic orders, lack of tunability is a serious drawback for the previously cited scientific investigations: it narrows accessible optical excitations down to few photon energies. Driving HHG with a mix of the fundamental wavelength and its second harmonic is a simple technique to produce both even and odd harmonics of the fundamental wavelength [16]. However, it only partially fills the gap between spectral lines, thereby not reaching a full tunability. Full tunability has been demonstrated for high harmonic orders, corresponding to extreme-ultraviolet (EUV) wavelengths, with techniques such as harmonic blueshifting depending on the generation geometry [17], control of the chirp of the drive laser [18, 19] or, in the picosecond range, tuning of a non-compressed narrow-band Ti:Sa drive source over its spectral range of amplification [20].

A more effective and straightforward solution to generate fully-tunable harmonics is to rely on a widely tunable drive source. It has been demonstrated in [21] for harmonics around 150 nm. In this paper, we extend this study to the whole ultraviolet spectral range. The novelty of our work stems from the unique qualities of the source that we used to drive HHG. This driving source is characterized by a large wavelength tunability from 1100 to 1900 nm, a mJ-level pulse energy and short pulse duration of the order of 20 fs. In these conditions the generation of few-femtosecond harmonic radiation is ensured. Our experiment and its implementation are described in the next section. Experimental results are then presented and discussed.

2. Experimental setup

The layout of the experiment is shown in Fig. 1. The parametric source is based on a Ti:Sapphire laser facility providing intense short pulses (tens of mJ energy; 60 fs duration), centered at a wavelength of 790 nm, with a repetition rate of 10 Hz. The output beam stems from difference frequency generation (DFG) of spectrally broadened pulses. The generated pulses are then amplified in a two-stage optical parametric amplifier (OPA), leading to the production of ~20-fs pulses with an energy up to 1.2 mJ, tunable from 1100 to 1900 nm. Tunability is achieved by rotating the crystals in the OPAs, thereby changing the phase-matching conditions.

The generation of the high-order harmonics of the near-IR driving pulses is achieved by focusing the laser beam on a jet of krypton gas, which ensures a better harmonic conversion efficiency than lighter gases such as argon, at the price of a lower cutoff frequency.

To cover the deep-ultraviolet (DUV) and EUV spectral regions two different spectrometers have been used. Harmonic emission in the DUV was analyzed through a normal-incidence Czerny-Turner scanning monochromator[1] equipped with a 2400-gr/mm $AlMgF_2$-coated grating. The monochromator selects a single harmonic or a spectral portion thereof. The photon flux at the exit slit of the monochromator is detected by a photomultiplier tube[2] with a tetraphenyl butadiene (TPB) phosphor photocathode to enhance the detection efficiency. Owing to the limited spectral range accessible to the grating, that has significant transmission for wavelengths above ~130 nm, only the harmonics ranging from the third to the eleventh order of the fundamental wavelength could be detected. The harmonic spectra at high resolution were obtained by scanning the grating, with a 300-μm slit aperture, giving a bandwidth of 0.4 nm.

The global response of the instrument (i.e., monochromator plus detector) has been absolutely calibrated using the facilities available at CNR-IFN and described in details in [23], in order to measure the DUV photon flux generated in the interaction region at the different harmonics. This was performed by tuning the monochromator to one of the harmonics and opening completely its slits. In such a way, the beam enters the monochromator without being clipped at the entrance slit and is diffracted by the grating. The harmonic of interest then exits the monochromator without being clipped at the output slit, and is detected by the photomultiplier. We verified that even with the slits completely open the different harmonics were clearly separated at the output.

The signal in the EUV was analyzed through a grazing-incidence flat-field spectrometer equipped with a 1200-gr/mm gold-coated grating and tuned in the 80–35 nm spectral region. The spectrum is acquired by a 40-mm-diameter microchannel plate intensifier with MgF_2 photocathode and phosphor screen optically coupled with a low-noise CCD camera. Also in this case, the global response of the instrument (i.e., grating and detector) has been absolutely calibrated, as described in detail in [24, 25]. Since the spectrometer works without an entrance slit, having the harmonics generation point as its input source, all the generated EUV photons enter the instrument and are diffracted onto the detector.

[1] McPherson model 218
[2] Hamamatsu model R1414

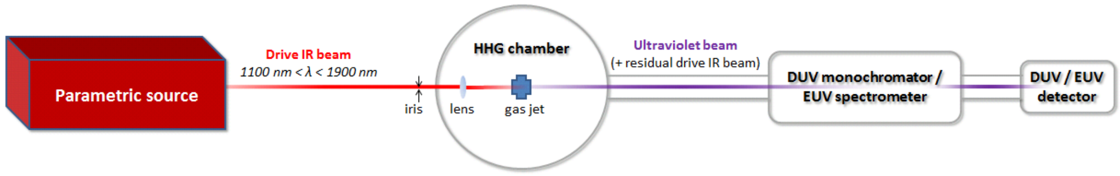

Figure 1: Layout of the experiment. The parametric source, whose comprehensive description can be found in [22], provides a tunable near-IR driving beam. It is focused with a lens of 15 cm focal length onto a krypton gas jet at a backing pressure of 3.5 bars. In order to attain the maximum harmonic yield for a given laser focal spot, the position of the jet with respect to the laser focus is optimized by carefully adjusting a manual x-y-z translation stage with a positioning precision of ± 0.01 mm. Moreover, an iris placed on the path of the driving beam allows to adjust size and intensity at the focus in order to optimize the harmonic generation efficiency. The gas is injected in the HHG chamber through an electromagnetic valve, synchronized with the laser beam, with a nozzle diameter of 0.5 mm operating at an opening time of 400 μs. The spectrum of the harmonic beam is acquired by two different detection systems: a monochromator in the DUV and a spectrometer in the EUV, with suitable detectors. Detection systems in both DUV and EUV regions have been calibrated so as to allow measurement of the harmonic absolute photon flux.

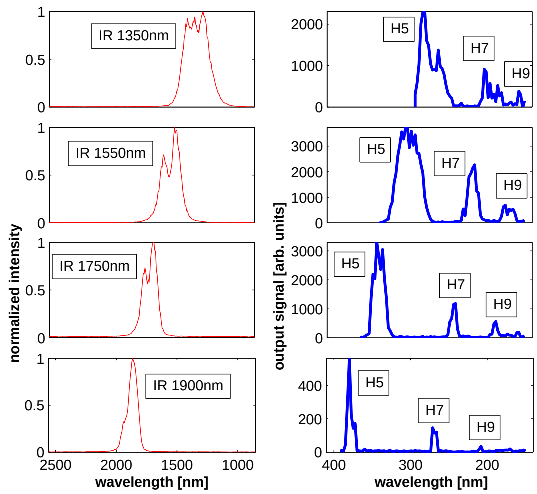

Figure 2: IR spectra (left side) and corresponding harmonic spectra (right side) in the 400–150 nm spectral region. H5, H7 and H9 stand respectively for the fifth, seventh and ninth harmonics of the driving IR beam.

3. Results and discussions

Deep-ultraviolet region

Figure 2 shows the spectral characterization from four sets of measurements, corresponding to four different wavelengths of the driving pulse: 1350, 1550, 1750, and 1900 nm. It is important to stress that the stability of the beam provided by the parametric source ensures a very good reproducibility of the measurements.

Below 150 nm, the efficiency of the DUV monochromator is dramatically low. Thereby, the analysis of harmonic spectra has been done only down to 150 nm. Since the third harmonics of the considered driving IR wavelengths are generally located in the visible, i.e., out of the monochromator range, harmonic orders from fifth to ninth have been analyzed. As expected at these relatively low orders, the signal quickly decreases with increasing harmonic order. Indeed, the intensity of harmonics before the plateau region is related to the probability of multiphoton ionization of the gas atoms [26]. Like the driving beam, harmonics have a large bandwidth (a few tens of nanometers), intrinsic to an ultrashort pulse source.

The overlap of the harmonic spectra shows a full tunability of the source in the DUV spectral region (Fig. 3). The range between 400 and 350 nm corresponds to either the fifth harmonic of a 1750–2000 nm fundamental beam or the third harmonic of a 1050–1200 nm fundamental beam. These wavelengths are the boundaries of the accessible spectral range of the used parametric source, so that in these regions the IR spectrum is less stable and moreover the beam energy is lower than in the 1350–1550 nm “peak region”. Hence harmonics in the 400–350 nm region are also less intense. The third harmonic of a 1050–1200 nm fundamental beam can be generated with better conversion efficiency in the frame of classical nonlinear optics in crystals [27].

Figure 4 clearly shows that when the driving wavelength ranges from 1350 to 1900 nm, as in these measurements, harmonic orders from fifth to eleventh completely cover the DUV spectral region. Furthermore, the third harmonic, not shown in Fig. 4, also allows tunability in the visible region. Obviously this overlap and thereby the tunability in the ultraviolet range improve at shorter wavelengths, where narrower IR tunability is thus sufficient.

The photon flux of the harmonics has been measured by fully opening the slits of the monochromator in order to get all the signal on the photomultiplier. The results are summarized in Table 1. Around 10^7 photons per

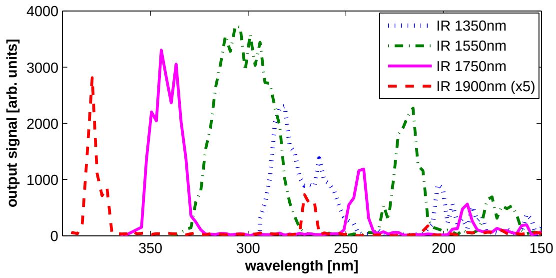

Figure 3: Overlap of the harmonic spectra for four drive IR wavelengths (1350, 1550, 1750 and 1900 nm). The harmonic spectrum resulting from the drive wavelength of 1900 nm has been vertically magnified (×5).

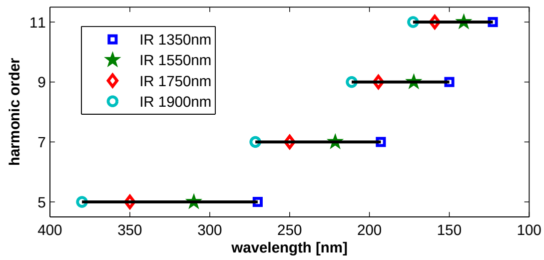

Figure 4: Tunability in the DUV. The lines represent the wavelength ranges that are covered by harmonic orders from fifth to evelenth, generated by drive wavelengths ranging from 1350 nm to 1900 nm.

Table 1: Absolute number of photons provided in the DUV spectral range for four different drive wavelengths. Left column: central wavelength of the drive beam; center column: wavelengths corresponding to the peak signal of the indicated harmonics; rigth column: measured photons/shot for each harmonic.

drive IR [nm]	harmonic [nm (order)]	photons/shot
1160	414 (3$^{\text{rd}}$)	2.0×10^8
	248 (5$^{\text{th}}$)	6.1×10^7
	177 (7$^{\text{th}}$)	3.6×10^7
1350	270 (5$^{\text{th}}$)	4.2×10^7
	196 (7$^{\text{th}}$)	1.8×10^7
	153 (9$^{\text{th}}$)	1.3×10^7
1450	285 (5$^{\text{th}}$)	6.1×10^7
	204 (7$^{\text{th}}$)	1.6×10^7
	161 (9$^{\text{th}}$)	8.2×10^6
1800	367 (5$^{\text{th}}$)	1.3×10^7
	262 (7$^{\text{th}}$)	7.0×10^6
	206 (9$^{\text{th}}$)	3.4×10^6

shot are generated in the DUV spectral region, corresponding to a beam energy of the order of 10 pJ. One sees that the higher the driving wavelength, the smaller the harmonic photon flux. Regarding the ninth harmonic of the driving laser for 1350, 1450 and 1800 nm drive wavelengths (λ_{IR}), the harmonic conversion efficiency scales as $\lambda_{\text{IR}}^{-6.34}$. This is in agreement with recent theoretical studies which show that the harmonic efficiency in the plateau region scales as λ_{IR}^{-6}, not as λ_{IR}^{-3}, as previously believed [28]. Moreover, in [29], the conversion efficiency of further plateau harmonics (from 78 to 39 nm) has recently been measured to be proportional to $\lambda_{\text{IR}}^{-6\pm1.1}$ in krypton. Although increasing the driving wavelength allows to extend the harmonic plateau [30], there is a penalty in terms of harmonic efficiency.

Extreme-ultraviolet region

The same procedure has been followed for the measurements performed in the EUV region, using the detection system described before. Harmonic spectra are reported in Fig. 5 for three different driving wavelengths (1350, 1450, 1550 nm) and their overlap in the 45–35 nm spectral range is shown in Fig. 6. Figure 7 shows that by varying the driving wavelength from 1350 nm to 1550 nm one attains the full tunability in the EUV, through relatively high-order harmonics. An interesting point is that one specific ultraviolet wavelength can be obtained from multiple drive wavelengths through different harmonic orders.

The absolute number of photons in the EUV is reported in Table 2 for harmonic orders 21, 29 and 35 of 1350, 1450 and 1550 nm driving wavelengths. Such a photon flux corresponds to an energy per harmonic per shot about two orders of magnitude smaller than in the DUV. This can be explained by the different nature of the harmonic generation process and of phase matching conditions in the DUV and EUV spectral regions.

Different strategies can be pursued to overcome this low photon flux. As a first possibility, one could design a more powerful parametric source [31]. A complementary strategy is the improvement of the HHG process in terms of tunability and conversion efficiency. In this respect, a promising technique that could be investigated is mixing the fundamental wavelength of the parametric source with either its second harmonic or with a standard powerful Ti:Sa laser source, as demonstrated in [32].

4. Conclusion

The full tunability of a femtosecond photon beam produced through HHG driven by a parametric source

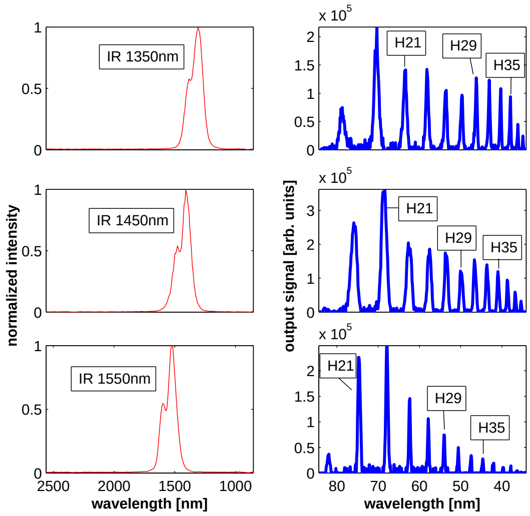

Figure 5: IR spectra (left side) and corresponding harmonic spectra (right side) in the 85–30 nm spectral region. H21, H29 and H35 stand respectively for the 21st, 29th, 35th harmonic order of the drive IR beam.

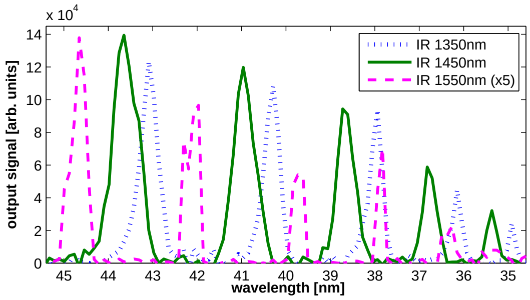

Figure 6: Overlap of the harmonic spectra in the range 45–35 nm for three drive IR wavelengths (1350, 1450 and 1550 nm). The harmonic spectrum resulting from the drive wavelength of 1550 nm has been vertically magnified (×5).

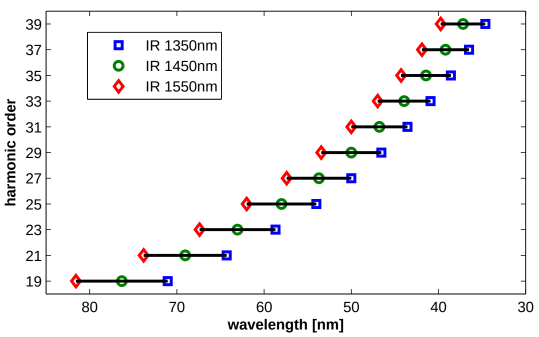

Figure 7: Tunability in the EUV. The lines represent the wavelength ranges that are covered by harmonic orders from number nineteen to thirty-nine, generated by drive wavelengths ranging from 1350 nm to 1550 nm.

Table 2: Absolute number of photons provided in the EUV spectral range for three different drive wavelengths. Left column: central wavelength of the drive beam; center column: wavelengths of harmonic orders 21, 29 and 35; right column: measured photons/shot for each harmonic.

drive IR [nm]	harmonics [nm]	photons/shot
1350	63	1.3×10^4
	46	5.8×10^3
	38	2.8×10^3
1450	69	4.4×10^4
	50	9.9×10^3
	41	4.7×10^3
1550	75	1.4×10^4
	54	3.1×10^3
	45	6.5×10^2

has been demonstrated in the whole ultraviolet spectral range. This source opens the way to novel scientific experiments. The main drawback comes from the relatively low harmonic conversion efficiency, resulting from a drive wavelength longer than in classic HHG setups. Increasing the harmonic photon flux would not only extend the range of possible scientific experiments, but also pave the way for the development of a tunable ultraviolet/soft X-ray source for seeding single-pass free-electron lasers, an application for which HHG-based sources have demonstrated to be attractive [33].

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